JOURNAL OF ENVIRONMENTAL SCIENCES XX (2017) XXX–XXX

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JES JOURNAL OF ENVIRONMENTAL SCIENCES www.jesc.ac.cn

Pollutants identification of ambient aerosols by two types of aerosol mass spectrometers over southeast coastal area, China

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ARTICLE INFO

Article history:
Received 15 February 2017
Revised 15 May 2017
Accepted 23 June 2017
Available online xxxx

Keywords:
Aerosol
Size distribution
Chemical composition
Aerosol mass spectrometer
Aerosol source

ABSTRACT

Two different aerosol mass spectrometers, an Aerodyne Aerosol Mass Spectrometer (AMS) and Single Particle Aerosol Mass Spectrometer (SPAMS) were deployed to identify the aerosol pollutants over Xiamen, representing the coastal urban area. Five obvious processes were classified during the whole observation period. Organics and sulfate were the dominant components in ambient aerosols over Xiamen. Most of the particles were in the size range of 0.2–1.0 μm, accounting for over 97% of the total particles measured by both instruments. Organics, as well as sulfate, measured by AMS were in good correlation with measured by SPAMS. However, high concentration of NH_4^+ was obtained by AMS, while extremely low value of NH_4^+ was detected by SPAMS. Contrarily, high particle number counts of NO_3^- and Cl^- were given by SPAMS while low concentrations of NO_3^- and Cl^- were measured by AMS. The variations of POA and SOA obtained from SPAMS during event 1 and event 2 were in accordance with the analysis of HOA and OOA given by AMS, suggesting that both of AMS and SPAMS can well identify the organic clusters of aerosol particles. Overestimate or underestimate of the aerosol sources and acidity would be present in some circumstances when the measurement results were used to analyze the aerosol properties, because of the detection loss of some species for both instruments.

Introduction

Particulate matter (PM) has become the primary pollutants in urban areas, since large quantities of aerosol particles were inevitably emitted into atmosphere with the rapid development of economic and energy consumptions (Yan et al., 2016; Yang et al., 2010). The importance of aerosols has received considerable attention throughout the world because of their impacts on air quality, human health (Buonanno et al., 2013; Brook et al., 2002), atmospheric visibility (Kanakidou et al., 2005) and global climate change (Prather, 2009; Alfarra et al., 2004). Different aerosol particle compositions would cause different health risks (Kang et al., 2004; Kim et al., 2008) and was also necessary to identify their sources and predict their effects on atmospheric processes.

Xiamen island located in the Southeast China is a well-known tourism coastal city on the Southeast coast of China and one of the Cleanest Cities of China (http://www.cnemc.cn/) with a sub-tropical climate under the influence of the Asian monsoon. In cold seasons of winter and spring, a northeasterly wind becomes the most prevailing wind direction. Air masses mainly come from northern China during wintertime. While in summertime, air masses from marine affect the local atmospheric quality. Therefore, aerosol particles in Xiamen area are not only related to anthropogenic activities, such as vehicular, coal combustion, and ship and industrial emissions,

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http://dx.doi.org/10.1016/j.jes.2017.06.030
1001-0742/© 2017 The Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences. Published by Elsevier B.V.

but also affected by the marine sources, such as sea salt particles, and sulfate and biological aerosols (Zhao et al., 2011; Yan et al., 2015; Zhang et al., 2011, 2012). In urbanized coastal zone, anthropogenic nitric acid reacts with sea salt to form coarse particles of sodium nitrate ($NaNO_3$), which increases the deposition velocities of particles and cleanses the atmosphere of acidic gaseous aerosol precursors. Moreover, the surface reaction often occurs between sulfuric acid and sodium chloride, producing sodium sulfate (Falkowska and Lewandowska, 2004; Chalbot et al., 2013). The biological components are primarily emitted into the atmosphere through a bubble bursting mechanism, usually as a part of mixed aerosol composed of other organic and inorganic compounds (Aller et al., 2005; Després et al., 2012; O'Dowd et al., 2004). In coastal area, the interaction between marine aerosols and anthropogenic particles was rather complicated and ambiguous. Therefore, it is important to investigate the chemical and physical properties of aerosol particles to identify their potential sources, formation mechanisms and atmospheric processes.

Recently, studies on aerosol properties over the coastal area of Xiamen including size distribution, source identification (Zhao et al., 2011; Zhang et al., 2012) have been present. However, offline filter sampling methods were conducted in previous studies with low time resolution. Long sampling time, 24 hr or even longer, was required to reach detection limits for offline filter analysis (Zhang et al., 2007). Since the physico-chemical process of aerosol particles in atmosphere occurred every moment, the physical and chemical characteristics of aerosol changed rapidly within a short time. Offline filter sampling methods were difficult to probe into the physico-chemical processes of aerosol particles in some special atmospheric events, because of a long sampling and analysis time. Therefore, high-time-resolution measurement was important and necessary to study the quick change of aerosol sources and compositions in ambient atmosphere.

Since online aerosol mass spectrometry was able to provide size and chemical information of aerosol particles in atmospheric environment with high-time-resolution (Sullivan et al., 2007; Spencer et al., 2007), It was used to characterize the aerosol chemical compositions, aerosol sources, mixed state and secondary aerosol formations, *etc.* In recent years, two types of aerosol mass spectrometers were used in atmospheric aerosol investigation, such as aerosol time-of-flight mass spectrometer (ATOFMS, Dall'Osto and Harrison, 2006; Dall'Osto and Harrison, 2012; Steve et al., 2012), and quadrupole aerosol mass spectrometer (Q-AMS, Jayne et al., 2000; Jimenez et al., 2003; Alfarra et al., 2004; Sun et al., 2010). Recently, a newly developed single particle mass spectrometer (SPAMS) was widely used in ambient aerosol researches (Li et al., 2014; Ma et al., 2016), which was able to provide the size and chemical information of a single particle which was important to identify aerosol sources. SPAMS and AMS have been extensively applied in atmospheric aerosol investigation. However, the comparisons of measurement results obtained by both instruments have not been elucidated. Different measurement results would be present when different types of measurement instruments were used to identify aerosol pollutants. This study aims to provide detailed comparisons of physical–chemical properties of atmospheric aerosol particles measured by SPAMS and AMS and understand impacts of measurement instruments on the atmospheric aerosol sources and pollutant identification. In this case, the investigation of aerosol pollutants over the coastal urban zone Xiamen, China was carried out using SPAMS and AMS simultaneously to demonstrate the discrepancy in aerosol measuring results.

1. Methods and data

1.1. Sampling site

The observation site (24°16′N, 118°05′E) was located on the platform of 10th Research Building about 45 m height above the sea level. This site is located in the seaside of southern Xiamen city, which represents the coastal urban district due to the impact of traffic, sea salt, residential, construction, ship and coal-fired emissions.

1.2. Instruments and measurements

The observation system included the sampling, control and measurement instruments. A $PM_{2.5}$ sampling cyclone (URG Corp., USA) was set to conduct intensive measurements. A Quadruple Aerosol Mass Spectrometer (AMS, Aerodyne Research Inc., USA) and a single particle mass spectrometry (SPAMS 0515, Guangzhou Hexin Analytical Instrument Co., Ltd., Guangzhou, China) were deployed to analyze the size-resolved chemical composition of aerosol particles simultaneously. A $PM_{2.5}$ sampler (MS310, USA) was employed at the same time to collect $PM_{2.5}$ samples at every 24 hr during the observation period. Particulates were collected on quartz fiber filters (Whatman, UK). Samples were stored in refrigerator at −20°C after sampling for later analysis. Eight samples were obtained during the observation period. Meteorological parameters were monitored by a weather transmitter WXT520 (Vaisala Co., Ltd., Finland).

1.3. Water-soluble inorganic ions analysis

A quarter of each blank and sample quartz fiber filter were cut into fine strips and dipped into 10 mL ultrapure water (18.2 MΩ/cm), and water-soluble inorganic ions from the samples were extracted by ultrasonic bath for 30 min. The extract solutions were filtered with a PTFE syringe filters (Pall Co., Ltd., USA). An ion chromatography system (ICs-2500, Dionex, USA) was used to determine the concentration of aerosol water-soluble ions. Field blank value was subtracted from the sample concentrations.

1.4. AMS and data analysis

Aerodyne Aerosol Mass Spectrometer (AMS) operation principles have been described in detail by Jayne et al. (2000) and Jimenez et al. (2003). In this study, the AMS was alternated between two modes, the mass spectrum (MS) mode and particle time-of-flight (PTOF) mode. The PTOF mode was a size distribution measurement mode for selected *m/z* settings of the AMS (Jimenez et al., 2003; Allan et al., 2003). The PTOF mode operation, signals of eleven *m/z*'s representative of NO_3^-

Please cite this article as: Yan, J., et al., Pollutants identification of ambient aerosols by two types of aerosol mass spectrometers over southeast coastal area, China, J. Environ. Sci. (2017), http://dx.doi.org/10.1016/j.jes.2017.06.030